

## **A Method for Optimizing Waste Management and Disposal Practices Using a Group-Based Uncertainty Model for the Analysis of Characterization Data – 13191**

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### **ABSTRACT**

It is a universal requirement for characterization of radioactive waste, that the consignor shall calculate and report a Total Measurement Uncertainty (TMU) value associated with each of the measured quantities such as nuclide activity. For Non-destructive Assay systems, the TMU analysis is typically performed on an individual container basis. However, in many cases, the waste consignor treats, transports, stores and disposes of containers in groups for example by overpacking smaller containers into a larger container or emplacing containers into groups for final disposal.

The current standard practice for container-group data analysis is usually to treat each container as independent and uncorrelated and use a simple summation / averaging method (or in some cases summation of TMU in quadrature) to define the overall characteristics and associated uncertainty in the container group.

In reality, many groups of containers are assayed on the same system, so there will be a large degree of co-dependence in the individual uncertainty elements. Many uncertainty terms may be significantly reduced when addressing issues such as the source position and variability in matrix contents over large populations. The systematic terms encompass both inherently “two-directional” random effects (e.g. variation of source position) and other terms that are “one-directional” i.e. designed to account for potential sources of bias.

An analysis has been performed with population groups of a variety of non-destructive assay platforms in order to define a quantitative mechanism for waste consignors to determine overall TMU for batches of containers that have been assayed on the same system.

## **INTRODUCTION**

All measurements have an inherent uncertainty associated with them. It is conventional to express the upper and lower bounds of reasonable knowledge of the variability on a given assay value (at a given confidence level) in terms of a “Total Measurement Uncertainty” (TMU) value. Standard practice is to report TMUs associated with each of the measured quantities such as nuclide activity or mass.

For Non-Destructive Assay systems, the TMU analysis is typically performed on an individual container basis. Thus, the challenge of defining and bounding the uncertainty is treated in isolation for each assay, addressing only the uncertainty associated with a single measurement on an individual container.

However, in many cases, waste consignors will treat, transport, store and dispose of their containers in groups. Examples of this practice are: overpacking smaller containers into a larger container, emplacing containers into groups for final disposal or grouping containers in specific sub-regions (e.g. cells or rooms) within an interim store or repository.

The current standard practice for container-group data analysis is usually to treat each container as independent and uncorrelated and use a simple summation / averaging method (or in some cases summation of TMU in quadrature) to define the overall characteristics and associated uncertainty in the container group.

In reality, many groups of containers are assayed in batches on the same system, so there will be a large degree of co-dependence in the individual uncertainty elements. Many uncertainty terms may be significantly reduced when addressing issues such as the source position and variability in matrix contents over large populations.

An improved model of the group TMU term provides a useful mechanism for consignors to improve their knowledge of the total radiological contents of waste consignments that may potentially generate cost savings in storage and treatment of waste.

## **GROUP TMU EVALUATION**

An analysis has been performed with population groups of a variety of non-destructive assay platforms in order to define a quantitative mechanism for waste consignors to determine overall TMU on a platform specific basis for groups of so-called ‘sibling’ containers that have been assayed over the same parent system over a given period of time.

### Portable Drum Assay System

Portable far-field High Resolution Gamma Spectroscopy (HRGS) assay is usually performed using a High Purity Germanium (HPGe) detector, a DigiDART™ multi-channel analyzer, and a laptop computer [1]. The system is deployed using a suitable universal cart such as the PSC TechniCART™. Figure 1 shows a typical arrangement.



Fig. 1. Example of Portable Non-Destructive Assay System using Far-Field High Resolution Gamma Spectroscopy

### Neutron Assay of Large Boxes

The SuperHENC system [2] (depicted in Figure 2) combines a high efficiency neutron assay with a high resolution gamma spectroscopy system in a single trailer for assay of drums and Standard Waste Boxes (SWBs) up to a maximum envelope of 138.4 cm wide by 94.0 cm high by 180.3 cm long. The neutron counter consists of arrays of He-3 detectors embedded in all six sides of the neutron counting chamber thus providing a high efficiency  $4\pi$  neutron detector. The gamma spectrometer consists of a single High Purity Germanium (HPGe) detector and a turntable to allow viewing different sides of the SWB. The turntable also serves as a scale for weighing the SWB during the gamma measurement.

The neutron assay chamber utilizes a six-sided arrangement of polyethylene moderated He-3 detectors. The detectors are filled to ten atmospheres pressure and have various active lengths. The exterior of the neutron chamber is clad with eight inches of polyethylene to shield against exterior neutron sources. Passive neutron coincidence counting and multiplicity techniques [3] are used to quantify the Pu-240 effective (Pu-240e) mass content of the waste container.

The SuperHENC measures the Pu-240e content using passive neutron coincidence counting and calculates the total plutonium content combining the Pu-240e value with either Acceptable Knowledge (AK) or direct gamma measurement for the plutonium isotopic mass fractions and other radionuclides present.



Fig. 2. Example of Standard Waste Box Assay System (SuperHENC)

The neutron counter uses the Add-A-Source (AAS) method [4] for matrix correction and normalization. The AAS is a Cf-252 source attached to a Teleflex™ cable that travels under the neutron assay chamber, stopping at six pre-selected positions. When not in use, the source is retracted from the chamber and stored in a polyethylene pig. The software calculates the measured response to the AAS, compares this to a reference count and calculates the matrix correction factor. The normalization is a simple and quick check on the empty neutron chamber counting efficiency compared to a reference initial source measurement.

The SuperHENC has, to date, been deployed at five locations in the United States Department of Energy (DOE) complex. A summary of the typical measurement uncertainty parameters [5] for operational SuperHENC systems is given in Table I.

Table I. Typical Measurement Uncertainties for SuperHENC

Measurement Uncertainty Component	Uncertainty Range	Uncertainty Type
Statistical	Variable (Pu-240e dependent)	Random
Matrix	3-18% (Matrix dependent)	Systematic
Source Geometry	2-14% (Matrix dependent)	Systematic
Calibration	1%	Bias
Background	0 - 15% (Pu-240e dependent)	Bias
Multiplication	0 – 1.5% (Pu-240e dependent)	Bias

## UNCERTAINTY MODEL

Quantification of TMU usually involves defining systematic and statistical terms. The statistical terms are random and generally observe well behaved variance for large population groups. The systematic terms encompass both inherently “two-directional” random effects (e.g. variation of source position) and other terms that are “one-directional” i.e. designed to account for potential sources of bias (e.g. uncertainty in the activity of the calibration source, or bias in the weigh scale). These terms will behave differently over large groups with the random terms converging to zero and the bias terms remaining constant (assuming the bias terms to be constant with respect to time).

Total Measurement Uncertainty  $\Delta_{TMU}$  is often expressed as a fraction of the reported nuclide activity by propagation of various error terms. A typical expression is given in Equation 1.

$$\Delta_{TMU} = \sqrt{\Delta_{Rand}^2 + \Delta_{Syst0}^2 + \Delta_{Bias0}^2} \quad (\text{Eq. 1})$$

Where  $\Delta_{Rand}$  is the fractional uncertainty associated with random effects (counting statistics),  $\Delta_{Syst0}$  is the fractional uncertainty associated with systematic effects (for a given container) that are unbiased and  $\Delta_{Bias0}$  is the fractional uncertainty associated with systematic effects (for a given container) that have a directional bias.

Examples of unbiased systematic effects are variation in matrix contents, variation in source geometry and background correction uncertainty. This type of effect will vary in magnitude in each assay (positive and negative) and over multiple measurements the effects will cancel out.

Examples of biased systematic effects are uncertainty in the calibration source activity and other calibration terms that will be constant in magnitude from one measurement to another (assuming the same calibration is used). This term does not cancel out when containers are grouped.

In the case where the activities of all containers in the group have approximately the same size, the fractional TMU for a group of N containers,  $\Delta_{TMUG}$  can be given by the expression shown in Equation 2.

$$\Delta_{TMUG} \cong \sqrt{\frac{\sum_{i=1}^N \sigma_{Rand,i}^2}{M^2} + \frac{\Delta_{Syst0}^2}{N} + \Delta_{Bias0}^2} \quad (\text{Eq. 2})$$

Where  $\Delta_{TMUG}$  is the fractional TMU in the summed activity (or mass) of the group,  $M$ , and  $\sigma_{Rand,i}$  is the random uncertainty in the  $i^{\text{th}}$  container.

In reality, the containers from a group tend to be randomly populated from a parent distribution

and will contain small, medium and high levels of activity. A log-normal distribution is often a good means to represent this population. In the log-normal distribution, it is the logarithm of the variable rather than the variable itself that is normally distributed. For example, two populations are shown in Figure 3 (drawn from experience with real waste containers).

Log-normal distributions are usually characteristic of processes that are dependent on the product of multiple independent variables. This type of distribution is widely encountered in physical and biological sciences. Log-normal distributions are characterized by their “location” ( $\mu$ ) and “scale” ( $\sigma$ ) parameters, so that if say mass ( $M$ ) is log-normally distributed then  $\ln(M)$  will have a normal distribution with mean of  $\mu$  and standard deviation  $\sigma$ .

The plutonium distribution (which is modeled in the standard waste boxes) has a location parameter,  $\mu$ , equal to 1.2 (equivalent to median Pu mass,  $e^\mu$ , of 3.32 g) and scale parameter  $\sigma$  of 1.2. A similar distribution is shown for uranium in drums where  $\mu = 2.30$  (i.e., median mass of 10 g) and  $\sigma = 1.9$ .

Note that if a distribution is log-normal then a normal distribution will be observed in the logarithmic term regardless of the logarithmic base used. In this example the natural logarithm was used, but these observations will hold equally true for, say, a base 10 logarithm with the only difference being that the respective location and scale parameters compared to natural logarithms would be multiplied by a simple factor, in the log-10 case by  $\ln(10)$ .

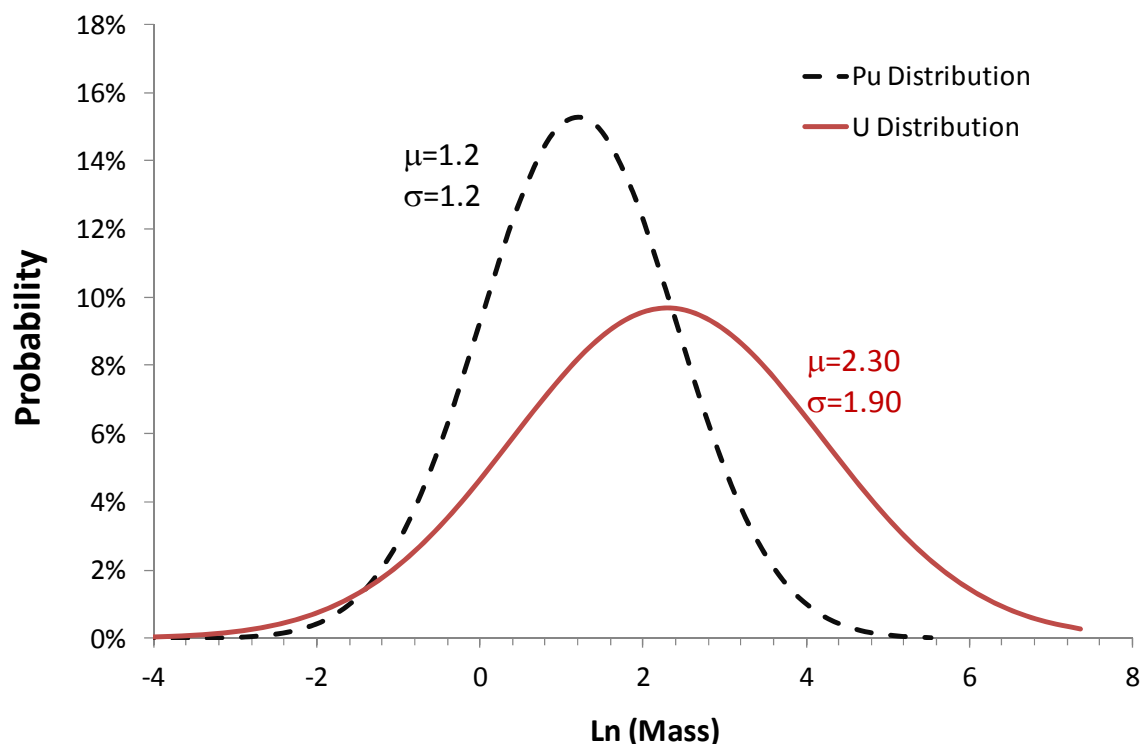


Fig. 3. Simulated Mass Distribution of Plutonium (for Standard Waste Boxes) and Uranium (for 208 Liter Drums)

In order to develop a general mathematical model of the group TMU term expressed in terms of the log-normal parameters and  $N$ , a computer simulation was created using Microsoft Excel 2007. In the simulation a Monte Carlo approach was used whereby a group of  $N$  containers was populated at random with mass ( $M$ ) values derived from the log-normal distribution using specified values of  $\mu$  and  $\sigma$ . The model was run with  $N = \{3, 10, 30, 100, 300, 1000\}$ ,  $\mu$  was sampled from values ranging from 1-9 and  $\sigma$  was sampled (independently from  $\mu$ ) in discrete values from 0 to 4.

For each iteration, the group fractional (unbiased) systematic TMU term  $\Delta_{SystG}$  was calculated for the population on  $N$  containers using Equation 3, which assumes that no covariance exists between the unbiased systematic uncertainty terms for the individual containers.

$$\Delta_{SystG} = \Delta_{Syst0} \frac{\sqrt{\sum_{i=1}^N M_i^2}}{M} \quad (\text{Eq. 3})$$

The results of the modeling are shown as the data points plotted in Figure 4. In this model the individual systematic term,  $\Delta_{Syst0}$  was set nominally to 20%. The plots show the group

systematic term plotted as a function of  $\sigma$  for various values of  $N$  (10, 100 and 1000 are shown). The model demonstrated that, for any given value of  $N$  and  $\sigma$ , a constant value was produced for the group systematic uncertainty term, when  $\mu$  was sampled over the range stated above. It was therefore concluded, that the group systematic uncertainty term is dependent only on  $N$  and  $\sigma$  and that there is no dependence on  $\mu$ .

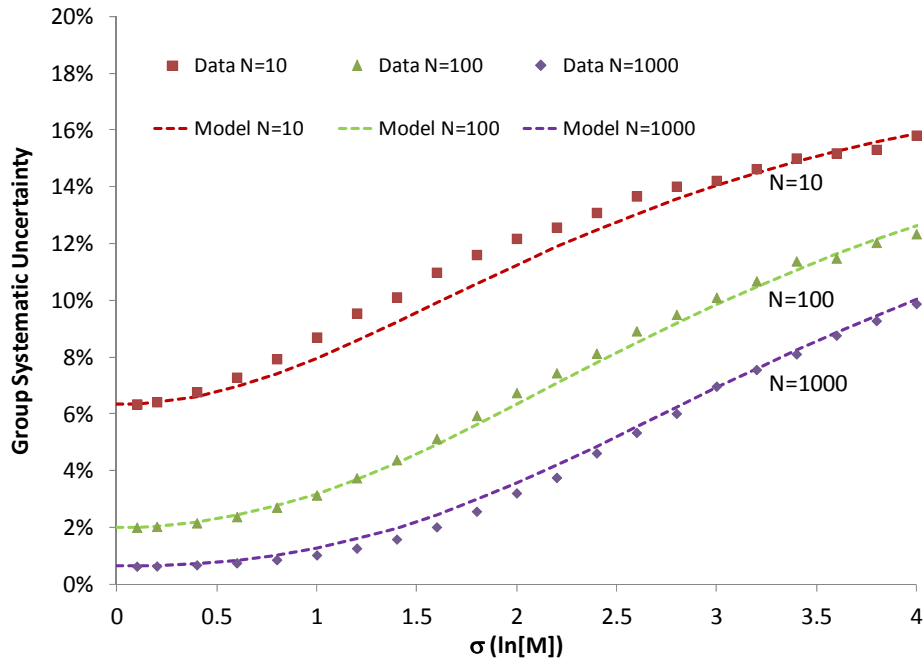


Fig. 4 Results of Monte Carlo Modeling of Group Systematic Uncertainty

It is useful to define the “systematic uncertainty ratio” ( $Y$ ) i.e. the ratio of the group systematic error ( $\Delta_{SystG}$ ) to the individual container systematic error ( $\Delta_{Syst0}$ ) as shown in Equation 4. The Monte Carlo derived curves demonstrate that  $Y$  can be represented to a reasonable approximation as a “power of  $N$ ” distribution with a simple functional dependence on  $\sigma$  as given in Equation 5.

$$Y = \frac{\Delta_{SystG}}{\Delta_{Syst0}} = N^{-\frac{Z}{2}} \quad (\text{Eq. 4})$$

$$\text{where } Z = \frac{1}{1 + \left(\frac{\sigma}{2}\right)^2} \quad (\text{Eq. 5})$$

It can readily be seen that in the case where  $\sigma \rightarrow 0$  then  $Y \rightarrow N^{-\frac{1}{2}}$  as expected, or in other words that where the population masses are evenly distributed then Equation 2 holds true. The curves that are generated by this model are illustrated in Figure 4, where it can be seen that the fit to the data



points is reasonable for values of N greater than 10.

Now returning to the general case, where bias uncertainty is present, for a group of  $N$  containers with an distribution of masses ( $M$ ) chosen from a parent that has a log-normal mass distribution with a scale parameter [i.e. standard deviation in  $\ln(M)$ ], of  $\sigma$ , then the TMU for the group (i.e. total TMU expressed as a fraction of total mass) can be represented by Equation 6.

$$\Delta_{TMUG} \cong \sqrt{(N^{-Z} \Delta_{Syst0}^2) + (\Delta_{Bias0}^2)} \quad (\text{Eq. 6})$$

In this expression it is assumed that the random uncertainty term diminishes to a negligible term for a large group of containers or in other words that the combined mass of the group is significantly greater than the system's lower limit of detection.

It can be seen that the first term on the right hand side of Equation 6 is equal to  $(Y^2 \Delta_{Syst0}^2)$ .

Thus, as stated above, when population masses are evenly distributed the first term becomes  $\frac{\Delta_{Syst0}^2}{N}$  and therefore, where the random error is negligible, Equation 2 reduces to Equation 6.

The Y term is plotted as a function of  $\sigma$  in Figure 5 for various values of N. In reality, the scale parameter  $\sigma$  will normally range between 0.7 and 3.2, the latter value representing a practical upper limit for waste.

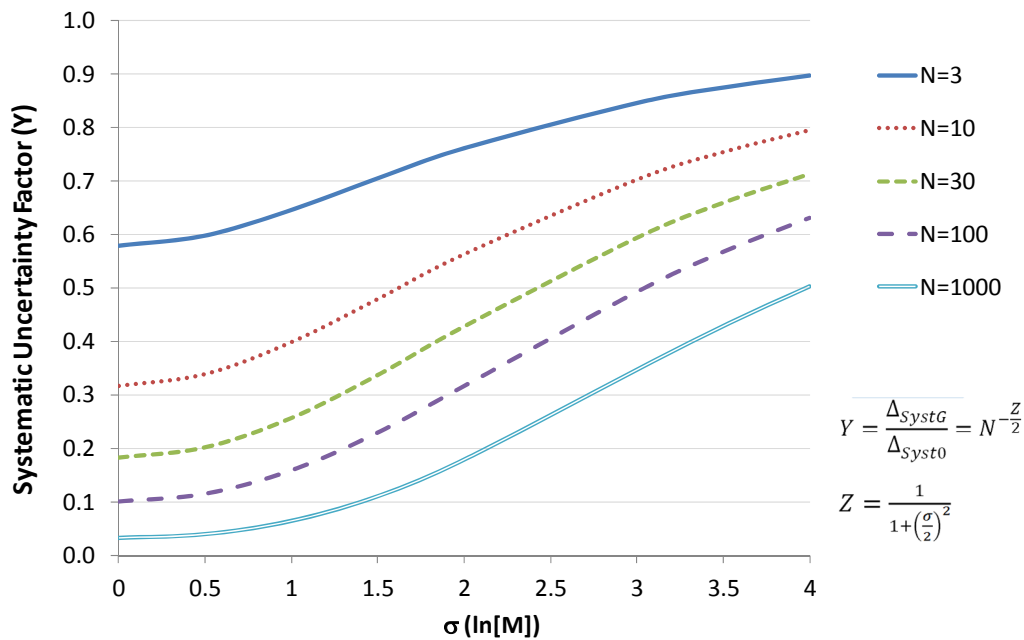


Fig. 5 Systematic Uncertainty Factor (Y) for Various Populations

### Uncertainty Model Parameters

For the uncertainty, the different NDA systems were assumed to have measurement uncertainty terms given in Table II. This is based in on the typical performance of similar instruments [1, 5].

TABLE II. Measurement Uncertainty Model Parameters

Measurement Uncertainty	Drum Assay (e.g. Portable Gamma)	Box Assay (e.g. SuperHENC)
Random	1 gram U= 10%	1 gram Pu = 3%
Systematic	40%	20%
Bias	10%	5%

### Drum Assay Group TMU analysis

The uranium mass in a hypothetical population of drums was sampled over 10,000 iterations using a mathematical model created in Microsoft Excel 2007 with a log-normal parent distribution. The “Load TMU” (for the Drum Assay System) was determined for (i) individual containers (ii) groups of 10 (iii) groups of 100 containers. The Load TMU is defined as the total TMU for the load divided the load total activity (or mass). For this assessment the groups were selected at random. Results are shown in Figure 6. Note how (i) random error in the single container diminishes with increasing mass, and (ii) the spread in the load uncertainty is reduced as the population size increases.

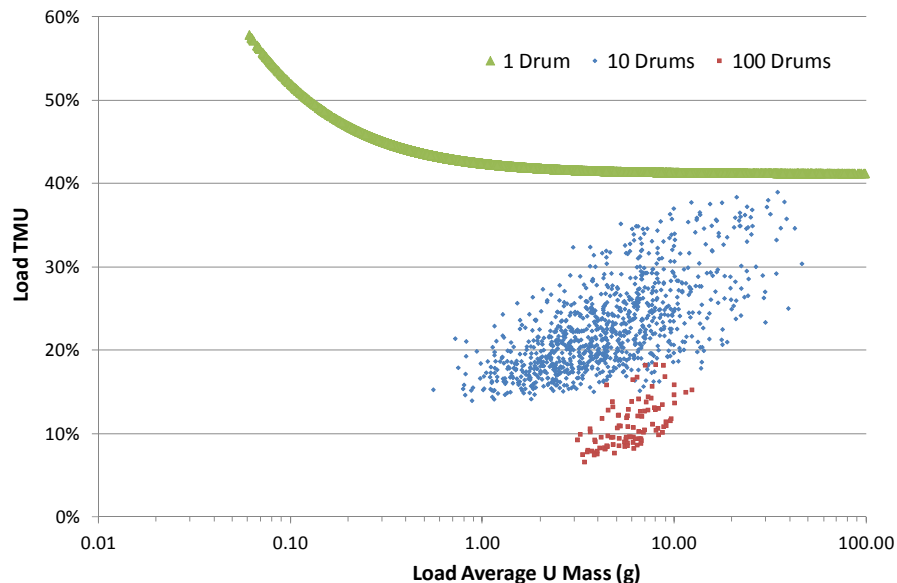


Fig. 6. Load TMU for Various Load Sizes of 208-liter Drums

### Box Assay Group TMU analysis

The plutonium population was sampled (with the mathematical model described above) over 10,000 iterations and a load TMU (for the Box Assay System) was determined for (i) individual containers (ii) groups of 10 (iii) groups of 100 containers. As with the drum analysis, the groups were selected at random. Results are shown in Figure 7.

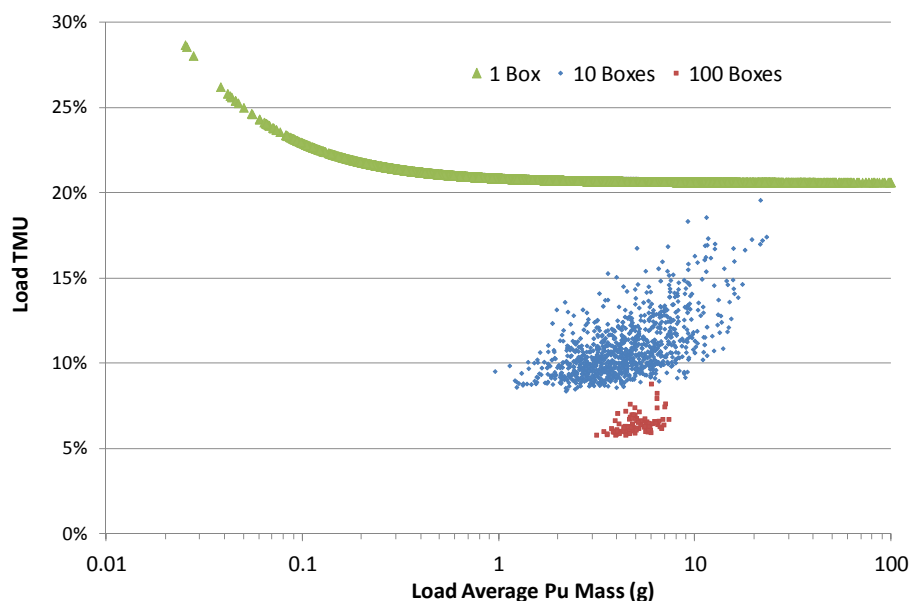


Fig. 7. TMU for Various Load Sizes of Standard Waste Boxes

### Group TMU Model Summary

Results of the group TMU model (defined in Equation 6) are shown in Table III. These estimates for the group TMU are consistent with the individual random iterations plotted in Figure 6 and Figure 7.

TABLE III. Group TMU Analysis

Number in Group	Drum Systematic Uncertainty Factor (Y)	Drum TMU	Box Systematic Uncertainty Factor (Y)	Box TMU
1	1.000	41.2%	1	20.6%
10	0.546	24.0%	0.429	9.9%
30	0.409	19.1%	0.286	7.6%
100	0.298	15.6%	0.184	6.2%
1000	0.163	11.9%	0.079	5.2%

## CONCLUSIONS

A model has been derived to allow the TMU term for a group of containers measured by Non-Destructive Assay to be estimated. This model requires only that the following information be available:

- Population statistics for the parent distribution (or a representative sample of the parent distribution) such that the standard deviation ( $\sigma$ ) in the logarithm distribution (also called the “scale parameter”) for the measured quantity may be calculated.
- An estimate of the total unbiased fractional systematic uncertainty component for each assay ( $\Delta_{\text{Syst0}}$ ) i.e. those components that will tend to cancel out over a large group (examples are matrix effects and source distribution terms).
- An estimate of the magnitude of the total biased systematic uncertainty component for each assay ( $\Delta_{\text{Bias0}}$ ) i.e. those components that are constant from one measurement to another (the usually includes the calibration uncertainty terms).

This model will be beneficial to waste consignors and experts involved in characterization of waste. Waste consignors can use the group based TMU model to assist in efficient and effective transport, sentencing or storage of waste (it may also be feasible to use the method retroactively on legacy data sets). This method will also be beneficial as guidance in the development of uncertainty reporting requirements for waste acceptance criteria.

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